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Review

Advances in the Application of Microcapsules as Carriers of Functional Compounds for Food Products

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Abstract: Natural bioactive compounds and living cells have been reported as promising products with beneficial properties to human health. The constant challenge regarding the use of these components is their easy degradation during processing and storage. However, their stability can be improved with the microencapsulation process, in which a compound sensitive to adverse environmental conditions is retained within a protective polymeric material. Microencapsulation is a widely used methodology for the preservation and stabilization of functional compounds for food, pharmaceutical, and cosmetic applications. The present review discusses advances in the production and application of microcapsules loaded with functional compounds in food products. The main methods for producing microcapsules, as well as the classes of functional compounds and wall materials used, are presented. Additionally, the release of compounds from loaded microcapsules in food matrices and in simulated gastrointestinal conditions is also assessed.

Keywords: microencapsulation methods; microcapsules; wall and core materials; stabilization of bioactive compounds; controlled release; incorporation in food matrix

1. Introduction

Research on microencapsulation began in the late 1930s. However, it was only in the 1950s that the microencapsulation process was first applied in industry, when the National Cash Register Company developed "carbonless carbon paper" using the coacervation technique. Currently, different industrial sectors such as pharmaceuticals, cosmetics, agriculture, textiles, and food use the process of microencapsulation for various purposes [1,2]. Moreover, as the encapsulation field is expanding rapidly, international groups and organizations such as the Bioencapsulation Research Group (http://bioencapsulation.net/) have been established. The aforementioned group has been active in the field of encapsulation since 1990, playing an important role in the development and improvement of encapsulation technologies and providing knowledge through conferences and training schools.

Concerning the food industry, the growing consumer interest in functional food products with nutritional quality, safety, improved shelf-life, and which can offer health benefits, has shifted the interest of researchers and companies to the use of artificial substances to natural bioactive compounds from fruits, vegetables, pulses, roots, and other plant sources [3,4]. However, many natural bioactive ingredients are unstable, being prone to oxidation, which is increased by exposure to light, and is also affected by heat, pH, and moisture content. As such, the food industry is interested in stabilization technologies for the preservation of the functional properties of bioactive materials during processing and storage, in modifying the physical properties of bioactive compounds to allow easier handling, in designing the release at the desired time and to specific targets, and in the increase of their bioavailability [5–7].

Microencapsulation is a process in which small particles or droplets of liquid are wrapped or coated by a polymeric material to produce small particles, which are called microcapsules or microspheres. The difference between them arises from their internal structure and morphology: microcapsules—the object of this review—are hollow internally possessing a reservoir system, while microspheres are dense matrix systems. This process allows the active ingredient, also named the core material, to be protected from adverse external environmental conditions by the coating, which is called the encapsulating agent or wall material [8–10]. The microparticles can exhibit different morphologies, being dependent on the properties of the core, wall material, and microencapsulation technique. Figure 1 shows a representation of the main types of microcapsules that can be formed, including multi-wall and single-wall systems, with single or multi-cores, with the core entrapped inside the capsule or within the wall, and with spherical or irregular shapes.

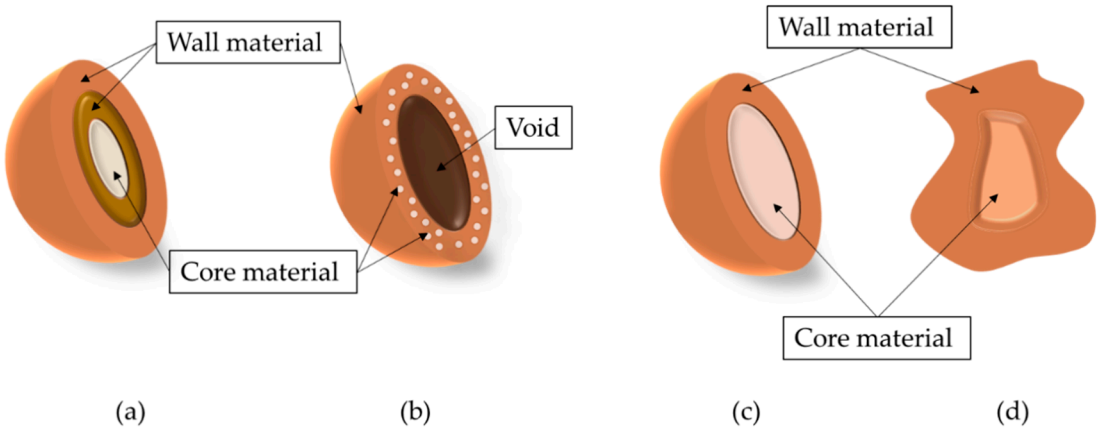

Figure 1. Main morphologies of loaded microcapsules. (**a**) multi-wall with single core; (**b**) single-wall with multi-core; (**c**) single-wall with single core; (**d**) irregular shape with single core and wall.

A large variety of works have been reported in the literature regarding the microencapsulation of natural bioactive compounds such as antioxidants present in aqueous extracts (e.g., phenolic compounds [11–15], carotenoids [16–21]), organic extracts, or essential oils [22,23], as well as living cells [24–28]. Microcapsules produced for the purpose of incorporation into food products must be formed by a food-grade wall material, and edible polymers such as maltodextrin, inulin, arabic gum, and starch, among others, have emerged as candidates [29].

The present work highlights the recent advances in stabilization strategies for the production of microcapsules loaded with bioactive natural materials for food applications. It includes the most commonly used encapsulation processes, the bioactive materials studied as core materials, and the food grade biopolymers used as wall materials. Additionally, attention is also focused on the stability of the encapsulated core materials upon incorporation in food products and the release properties in the gastrointestinal tract after ingestion in order to envisage their potential bioavailability (Figure 2).

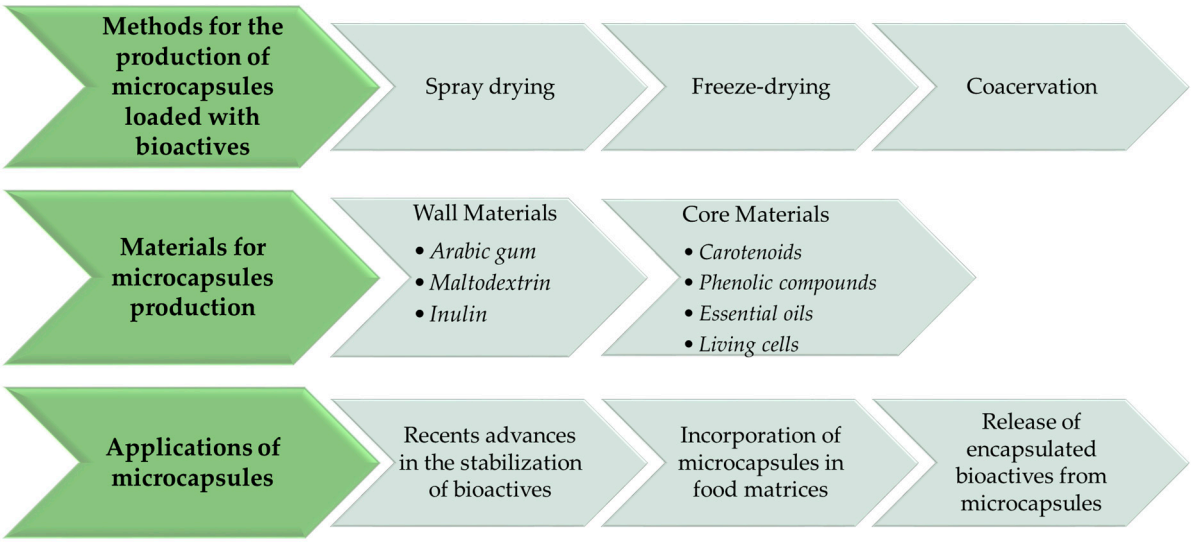

Figure 2. Summary of topics addressed in this paper.

2. Methods for the Production of Microcapsules Loaded with Bioactive Compounds

Several methods for the encapsulation of bioactive compounds have been studied, such as spray drying, coacervation, freeze drying, electrospraying, ionic gelation, and fluidized bed coating [30]. Table 1 shows an overview of the encapsulation systems referred recently in the literature, indicating the methods, the wall materials, and the bioactive compounds used.

Table 1. Encapsulation systems of bioactive compounds for food applications.

Wall Material	Core Material	Core Material Source	Main Properties/Applications Studied	Refs.
		Spray Drying		
Arabic gum Gelatin Maltodextrin	Anthocyanins	Barberry extract	Stabilization of active ingredient	[31]
Arabic gum Maltodextrin	Anthocyanins Carotenoids	Tamarillo	Stabilization of active ingredient Storage stability	[32]
Arabic gum Maltodextrin Whey protein	Carotenoids	Carrot	Stabilization of active ingredient	[33]
Arabic gum Whey protein	Carotenoids	Gac oil	Storage stability Incorporation in food matrix	[34]
Maltodextrin	Carotenoids	Mango Banana Tamarillo	Stabilization of active ingredient Storage stability	[16]
Maltodextrin Sodium caseinate	Carotenoids	Red palm oil	Stabilization of active ingredient	[17]
Arabic gum Soy protein	Carotenoids	Tomato oleoresin	Stabilization of active ingredient Storage stability Controlled release of bioactives from microcapsules	[19]
Arabic gum	Carotenoids	β-carotene	Stabilization of active ingredient	[35]
Alginate Chitosan Inulin	Essential oil	Coriander	Controlled release of bioactives from microcapsules	[23]
Inulin	Essential oil	Oregano	Controlled release of bioactives from microcapsules	[36]

Table 1. *Cont.*

Wall Material	Core Material	Core Material Source	Main Properties/Applications Studied	Refs.
Maltodextrin Whey protein	Essential oil	Lime	Controlled release of bioactives from microcapsules	[22]
Maltodextrin	Lutein	Marigold flowers	Stabilization of active ingredient	[18]
Maltodextrin	Phenolic compounds	Laurel infusions	Controlled release of bioactives from microcapsules	[37]
Arabic gum Maltodextrin	Phenolic compounds	*Renealmia alpinia*	Stabilization of active ingredient Storage stability	[14]
Arabic gum Maltodextrin Skimmed milk Whey protein	Phenolic compounds	Pomegranate peels	Stabilization of active ingredient Incorporation in food matrix	[13]
Arabic gum Maltodextrin β-Cyclodextrin Chitosan Gelatin	Phenolic compounds	Plum	Storage stability	[38]
Maltodextrin	Phenolic compounds	Cinnamon infusions	Stabilization of active ingredient Controlled release of bioactives from microcapsules	[39]
Maltodextrin	Phenolic compounds	Averrhoa carambola pomace	*In vitro* simulated gastrointestinal digestion release of bioactives from microcapsules Storage stability	[40]
Maltodextrin Pea protein Whey protein	Phenolic compounds	Grape marc	*In vitro* simulated gastrointestinal digestion release of bioactives from microcapsules	[41]
Whey protein	Polyphenols	Vanilla	Stabilization of active ingredient Storage stability	[42]
Arabic gum Inulin	Probiotic bacteria	*Lactobacillus acidophilus*	Storage stability *In vitro* simulated gastrointestinal digestion release of probiotics from microcapsules	[24]
Arabic gum Maltodextrin Modified starch Whey protein	Probiotic bacteria	*Saccharomyces cerevisiae*	*In vitro* simulated gastric digestion release of probiotics from microcapsules	[43]
Arabic gum High maize starch Maltodextrin	Probiotic bacteria	*Lactobacillus acidophilus*	Storage stability	[44]
Goat's milk Inulin Oligofructose	Probiotic bacteria	*Bifidobacterium*	*In vitro* simulated gastrointestinal digestion release of the probiotics from microcapsules	[45]
Inulin Maltodextrin	Probiotic bacteria	*Bifidobacterium*	Stabilization of active ingredient Storage stability	[25]
Maltodextrin Skim milk Trehalose	Probiotic bacteria	*Lactobacillus casei*	*In vitro* simulated gastrointestinal digestion release of the probiotics from microcapsules	[46]
Arabic gum	Vitamin A	Retinol	Storage stability Controlled release of bioactives from microcapsules	[47]

Table 1. *Cont.*

Wall Material	Core Material	Core Material Source	Main Properties/Applications Studied	Refs.
		Freeze-drying		
Maltodextrin	Anthocyanins	Blackberry pulp pomace	Stabilization of active ingredient	[12]
Arabic gum Whey protein	Anthocyanins	Sour cherries	*In vitro* simulated gastrointestinal digestion release of bioactives from microcapsules Incorporation in food matrix	[15]
Maltodextrin	Phenolic compounds	Averrhoa carambola pomace	*In vitro* simulated gastrointestinal digestion release of bioactives from microcapsules	[40]
Maltodextrin β-cyclodextrin	Polyphenols	Green tea	Incorporation in food	[48]
Denatured whey protein Fructooligosaccharide Sodium alginateWhey protein	Probiotic bacteria	*Lactobacillus plantarum*	Stabilization of active ingredient Storage stability	[49]
Inulin Persian gum Whey protein	Probiotic bacteria	*Lactobacillus rhamnosus*	*In vitro* simulated gastrointestinal digestion release of probiotics from microcapsules Storage stability	[26]
Maltodextrin	Probiotic bacteria	*Saccharomyces cerevisiae*	Controlled release of bacteria from microcapsules Storage stability	[27]
		Coacervation		
Arabic gum	Anthocyanins	Black raspberry	Stabilization of active ingredient Storage stability	[50]
Arabic gum Whey protein	Astaxanthins	*Haematococcus pluvialis*	Storage stability *In vitro* and *in vivo* simulated gastrointestinal digestion release of bioactives from microcapsules	[51]
Arabic gum Whey protein	Carotenoids	Sea buckthorn	Stabilization of active ingredient	[52]
Arabic gum Whey protein	Carotenoids	Sea buckthorn	Incorporation in food matrix	[21]
Chitosan Pectin Xanthan gum	Carotenoids	Palm oil	Incorporation in food matrix *In vitro* simulated gastrointestinal digestion release of bioactives from food matrix	[53]
Carboxymethylcellulose Chitosan Sodium tripolyphosphate	Carotenoids	Palm oil Soybean oil with β-carotene	Incorporation in food matrix *In vitro* simulated gastrointestinal digestion release of bioactives from microcapsules and food matrix	[54]
Arabic gum Gelatin	Phenolic compounds	Echium oil	Stabilization of active ingredient	[55]
Arabic gum Gelatin	Phenolic compounds	Broccoli	Stabilization of active ingredient	[56]
Arabic gum Gelatin	Proanthocyanidin	Cinnamon	Stabilization of active ingredient	[11]
Arabic gum Gelatin	Probiotic bacteria	*Bifidobacterium*	*In vitro* simulated gastrointestinal digestion release of probiotics from microcapsules Storage stability	[57]

Table 1. *Cont.*

Wall Material	Core Material	Core Material Source	Main Properties/Applications Studied	Refs.
Arabic gum Whey protein	Probiotic bacteria	*Lactobacillus paracasei* *Lactobacillus paraplantarum*	Stabilization of active ingredient *In vitro* simulated gastrointestinal digestion release of probiotics from microcapsules	[28]
Arabic gum Gelatin	Xylitol	Commercial	Stabilization of active ingredient Controlled release of bioactives from microcapsules	[58]

Spray drying is one of the oldest techniques used for the production of microcaparticles, in which dehydrated products in the form of fine powders are obtained. Nowadays, this technique is widely used in the food industry due to its low production costs, simplicity, ease of scaling up, and ability to produce microparticles with good properties for several uses. Moreover, water removal by spray drying ensures the microbiological quality and facilitates the transport, dosing, and storage of the obtained products [59,60]. The main drawback of the spray drying technology is the type of wall materials that can be used: as most formulations used by the food industry in this process are aqueous-based, the wall material used must have a good water solubility and simultaneously impart a suitable viscosity in order to enable the drying process [2,61].

The microencapsulation process by spray-drying is comprised of the following steps: feed preparation (may be a solution, a suspension, or emulsion, containing the wall and core materials), atomization of the feed in small droplets, rapid drying of liquid droplets in contact with a stream of hot gas (e.g., air) resulting in the instantaneous formation of the microparticles in the form of powder, and finally powder recovery. The temperature of the drying inlet air used is usually between 110 and 220 °C, and the exposure time of the feed solution at these high temperatures is only a few milliseconds. The temperature inside the microparticles, where the core material is present, is normally below 80 °C, which helps to minimize the thermal degradation of the material [1,2,62].

The properties of the resulting powder are influenced by the type of atomizer, feed physicochemical properties, drying air inlet and outlet temperatures, as well as the type and concentration of wall and core materials. When microcapsules are produced with hydrophobic active ingredients dispersed in feed emulsions, the microencapsulated bioactives are normally distributed within the thin wall and/or in the inner space. However, microencapsulated bioactives are more likely to be distributed within the thin wall if they are completely soluble in the feed solution along with the wall materials. Microcapsules usually show a spherical shape, with a low average particle size ranging from 5 to 50 μm, and have a smooth outer surface or exhibit the formation of teeth or concavities with an irregular shape [1,63,64].

Microencapsulation by freeze-drying is a process in which the feed containing the wall and core materials is frozen at temperatures below −40 °C and dried by sublimation under low pressure. The freeze-drying technique is simple and suitable for the microencapsulation of biological materials sensitive to heat and oxygen due to the application of low temperatures and removal of oxygen during the drying process. Different active ingredients such as phenolic compounds, carotenoids, and probiotic bacteria have been stabilized by the freeze-drying method [64,65]. However, this technique has some constraints, such as high energy consumption and operational cost due to the long processing time (above 20 h). In comparison to spray drying, the freeze-drying method can be up to 50 times more expensive. Furthermore, the dried material has a porous structure which exposes the active ingredient encapsulated to the atmosphere, thus offering low protection for a prolonged shelf life of that compound [20,61,66].

Coacervation is a process in which the separation of two liquid phases in an aqueous colloidal solution occurs. One phase is rich in colloid/polymer (coacervate) and the other phase, called the

equilibrium solution, is poor in polymer. This phase separation may be induced by changing the ionic strength, temperature, pH, or solubility of the dissolving medium [20,62].

The process of coacervation in aqueous phase is classified as simple coacervation when it is induced in a system where the wall material is a single polymer, or complex coacervation that is characterized by the interaction between oppositely charged wall materials, usually a protein and a polysaccharide, leading to a phase separation due to the electrostatic attraction between macromolecules. Several polymers have been used to produce complex coacervates, such as gelatin, arabic gum, whey protein isolate, chitosan, pectin, pea protein, and alginate, among others (Table 1) [20,62,67]. Complex coacervation has been used for the microencapsulation of different unstable active ingredients such as carotenoids [21,52,54], oils [53,55], phenolic compounds [50,51,56], and probiotic bacteria [28,57] (Table 1). Four major steps are involved in this encapsulation process: emulsification, coacervation itself, gelation, and hardening. In some cases, crosslinking agents, such as transglutaminase, calcium ions, or tripolyphosphate, are used to increase the strength of the capsule wall during the gelation and hardening steps [68].

Microcapsules produced by complex coacervation can present various morphologies and sizes (ranging from 5 to 200 μm) depending on changes in pH, ionic concentration, emulsion formation method, as well as the type and concentration of wall materials and bioactive compounds. Normally, capsules obtained at low homogenization rate during the emulsification process show a mononuclear morphology, that is, a single droplet with the active ingredient is surrounded by the shell. However, when they are formed at high homogenization rate they are more likely to have a multinucleated morphology, i.e., multiple small droplets of core materials surrounded by a bigger shell [20,68].

3. Wall Materials

Wall materials have different chemical structures and physicochemical properties that influence the efficiency of the microencapsulation process. As such, their correct selection is an important step due to their direct effect on the stability of the microcapsules, efficiency of core material retention, and shelf life [66,69]. The wall material must have suitable rheological properties at high concentrations and the ability to emulsify the active material, stabilize the produced emulsions, and keep the core material within its structure during processing or storage [66,70]. It is also quite important the purpose for which the microcapsules are produced, such as easier dosage and increased stability during storage, to mask undesirable taste of the core material, or the controlled release on a specific site in the human gastrointestinal tract [71].

Various types of wall materials have been used for the production of microcapsules, such as polysaccharides (e.g., starches, carrageenan, maltodextrins, and arabic gum), lipids (e.g., stearic acid, mono- and diglycerides), proteins (e.g., albumins, pea protein, gelatin, and casein), and their mixtures (Table 1). Of these, maltodextrin and arabic gum are two of the most commonly applied [31,72,73].

Arabic gum (*gum acacia*) is a complex exudate of acacia trees, of which there are many species distributed over tropical and subtropical regions. The most important growing areas for species that give the best gum are Sudan and Nigeria. Arabic gum is commonly used as a purified spray-dried powder [31,74]. It is a highly branched edible biopolymer and is chemically described as a mixture composed of carbohydrates consisting of D-glucuronic acid, L-rhamnose, D-galactose, 4-O-methyl-D-glucuronic acid, and L-arabinose, including approximately 2% protein [47,75]. Due to its highly ramified structure, it is easily dispersed when stirred in water in concentrations of up to 50%. This gum is used in the fabrication of microcapsules mainly due to its low viscosity at high concentrations, good emulsifying properties that are especially interesting to the microencapsulation of oils and flavors [31,73], compatibility with high sugar concentrations, subtle taste, and high oxidative stability [73,74].

Other applications of arabic gum in food are as a component of the glaze or coating of pan-coated candies in confections with high sugar and low water contents such as caramels, toffees, and jujubes. In confections, it prevents sucrose crystallization and emulsifies and distributes fatty components [74].

Nonetheless, arabic gum has some disadvantages, such as high cost and limited supply, as it is only produced in regions that are subject to unpredictable climate variations [75,76].

Maltodextrins are produced from starch hydrolysis with values of dextrose equivalent (DE) below 20. The degree of hydrolytic conversion of starch, expressed as DE value, is the criterion for the classification and characterization of hydrolysates. The higher the value of DE, the higher the degree of hydrolysis the product was subjected to. For DE values above 20, the hydrolysate is termed syrup solids or dextrin [77,78].

Maltodextrin has been widely used in the microencapsulation of bioactive compounds due to its satisfactory performance, low relative cost, and neutral taste and aroma. It is characterized by high solubility in water, low viscosity at high concentrations, film forming capacity, and good protection against the oxidation of core materials. On the other hand, the main disadvantage of the use of maltodextrin in the microencapsulation process is the low emulsifying ability. Thus, to form stable emulsions, mixing with other wall materials such as arabic gum, modified starch, and proteins is encouraged [69,73,79].

Mahdavi et al. [31] evaluated the influence of maltodextrin alone and mixed with gelatin or arabic gum when used as wall material in the microencapsulation of natural anthocyanins by spray drying. The authors concluded that the mixture of maltodextrin with arabic gum imparted a better protection to anthocyanin pigments and presented a higher encapsulation efficiency. In terms of particle morphology, that of maltodextrin/arabic gum was smooth and had fewer agglomerates and concavities on their surface when compared to maltodextrin and maltodextrin/gelatin microcapsules.

Inulin is another promising material for use in encapsulation. This polymer belongs to the group of fructans composed mostly of β(2-1) linked fructosyl-fructose units wherein each fructose chain is generally terminated with an α(2-1) linked glucose moiety [80–82]. The chain lengths of these fructans range from 2 to 60 units, with an average DP of 12. DP influences important properties, such as solubility, thermal stability, sweetness power, and prebiotic activity [83,84]. Standard chicory inulin has an average DP of about 10 to 12. Long-chain (or high performance) chicory inulin, from which the lower DP fraction has been physically removed, with an average DP of about 25, is also available [85].

Inulin has attracted attention from various researchers and industries due to reports of its many benefits, such as the nutrition of beneficial intestinal bacteria, the reduction of the risk of gastrointestinal diseases, the regulation of blood glucose, the promotion of mineral absorption, the improvement of the immune system, the prevention of osteoporosis, and the reduction of the risk of obesity and cholesterol level [86–88].

Inulin is not digested by the human digestive system, which does not have enzymes to separate the β (2-1) bonds in the fructose backbone. Therefore, most dietary inulin has the ability to stimulate the proliferation of specific bacteria in the lower colon (*bifidobacterium* and *lactobacillus*), thus reducing the number of harmful species such as *Escherichia coli* and *Clostridium* spp. [81,89].

The main applications of inulin are related to its technological properties, such as a substitute for sugars and fats in low-calorie foods, as a thickener, emulsifier, and gelling agent, and it may constitute a potential auxiliary agent for drying processes [80,84]. In the field of bioactives encapsulation, the use of inulin as wall material enables its health benefits to be harnessed, and at the same time it allows the release of bioactive compounds in the colon due to its resistance to pH variations in the gastrointestinal tract [23,90].

4. Core Materials

The core materials, which are also called coated material and actives, among other names, can be pure active substances or mixtures of high-added-value components extracted from plant leaves, flowers, stems, fruits, vegetables, or several agricultural residues. Examples of such materials are natural antioxidant bioactive compounds (carotenoids, phenolic compounds), vitamins, flavors, enzymes, and microbial cells (e.g., probiotic bacteria) [91–93] (Table 1).

Carotenoids, the most studied of the natural food pigments, are widely studied due to their beneficial effects on human health and their color. Some of the main benefits of carotenoids are the prevention of many diseases such as cancer, heart and vascular diseases, cataracts, hypertension, age-related macular degeneration, and diabetes, among others. Additionally, carotenoids also have the functionality of enhancing intercellular communication and immune system activation. Carotenoids are divided into two classes based on functional group: (1) xanthophylls, molecules containing oxygen such as lutein, zeaxanthin, and β-cryptoxanthin; and (2) carotenes, non-oxygenated molecules such as α-carotene, β-carotene, and lycopene. [94,95]. These are lipophilic pigments responsible for imparting the yellow, orange, red, and green color in many foods such as carrots, sweet potato, pumpkin, broccoli, and spinach (α-carotene and β-carotene), tomatoes, watermelon, and guava (lycopene), mandarin, orange, papaya, and egg yolks (β-Cryptoxanthin), and leafy greens such as spinach, collard, and kale (lutein and zeaxanthin) [94].

Phenolic compounds have been considered as promising bioactive molecules for the pharmaceutical and food industries. They constitute one of the most numerous groups of plant secondary metabolites. This group is constituted by molecules of different molecular weights and chemical structures, such as flavonoids, phenolic acids, stilbenes, tannis, and coumarins. Phenolic compounds are antioxidant, anti-inflammatory, and/or as antimicrobial agents. Therefore, the ingestion of polyphenol-rich food products may reduce the risk of developing cancer, cardiovascular diseases, diabetes, and osteoporosis. Fruits (e.g., grapefruit, lemon, blueberry, and strawberry), vegetables (e.g., potato, spinach, asparagus, artichoke, and lettuce), beverages (e.g., coffee, red wine, black and green teas, and orange and grapefruit juices), and herbs (e.g., basil, parsley, thyme, and mint) are examples of sources of phenolic compounds [40,96]. Foods enriched in these compounds show a delay in the formation of toxic oxidizing products and a prolonged shelf life [97].

Essential oils are complex mixtures of volatile lipophilic substances characterized by a strong flavor. They are plant secondary metabolites that can be synthesized by the whole plant (e.g., mint, lavender) or stored in certain parts of the plant, such as flowers (e.g., chamomile, bergamot), leaves (e.g., eucalyptus, basil, and lemongrass), rhizomes (e.g., turmeric, ginger), or seeds (e.g., nutmeg, cloves). They are commonly used in the cosmetic, pharmaceutical, and food industries due to their flavor or biological activities (e.g., antioxidant and antimicrobial properties). However, large losses of essential oil active compounds are common during the handling and storage due to their extreme sensitivity to oxidation, resulting in losses of their valuable properties [29,98].

Probiotic bacteria are a group of bacteria that when present in sufficient quantity may confer health benefits to the host. Human health benefits associated with probiotics include efficacy in the treatment of various intestinal disorders, inhibition of pathogenic microorganisms, improved tolerance to lactose, prevention of some cancers, reduction of total cholesterol, improvement of the immune system, and increase of the intestinal microbiota. However, probiotics must survive in the digestive tract, that is, they must be able to pass through the acidic conditions (pH = 2) of the gastric environment and reach the intestine in sufficient quantities to allow their colonization and proliferation. Additionally, the viability of probiotics is affected by other factors than pH, such as hydrogen peroxide, oxygen, and storage temperature, which may limit their effectiveness in most functional foods. Thus, microencapsulation of probiotics is an alternative to protect them against these adverse conditions. The probiotic microorganisms most used in the food industry and in microencapsulation processes are *Lactobacillus* and *Bifidobacteria* strains [67,99,100].

5. Recent Advances in the Stabilization of Bioactive Compounds

Over the past five years, researchers have shown great interest in using spray-drying, freeze-drying, and coacervation techniques using different wall and core materials. Wall materials, such as maltodextrin and arabic gum, and core materials, such as probiotic bacteria, phenolic compounds, and carotenoids were some of the most widely used materials (Table 1).

The stabilization of carotenoids from fruit juices (carrot and tamarillo) was performed by spray-drying in studies comparing the influence of different wall materials (arabic gum and maltodextrin) on chemical and physicochemical properties of the microcapsules [32,33]. The authors observed that arabic gum particles presented greater carotenoid retention and encapsulation efficiency when compared to the maltodextrin ones. Furthermore, Ramakrishnan et al. [32] also analyzed the storage stability of the microcapsules in relation to their antioxidant activity and the amount of carotenoids retained under different storage conditions. It was observed that powders produced with arabic gum stored at 25 °C for 24 days had a higher rate of carotenoid degradation than powders produced with maltodextrin. This fact was attributed to a higher rate of water activity at the beginning of the storage period, being more susceptible to degradation of the compounds and loss of antioxidant activity.

Another microencapsulation technique that has been used to stabilize carotenoids is complex coacervation. Mihalcea et al. [52] and Ursache et al. [21] studied the stabilization of carotenoids from sea buckthorn by microencapsulation using whey protein isolate as wall material. In both works, the authors suggest that the wall material used is a capable matrix for the encapsulation of carotenoids, presenting encapsulation efficiency varying between 41% and 56%. Also, laser confocal scanning microscopy confirmed that carotenoids were within the coacervates produced. Overall, the authors recommend that the microcapsules produced in the study have potential for use in the food industry as a natural source of antioxidants.

Yinbin et al. [38] evaluated the effect of different combinations of wall materials (maltodextrin/arabic gum, maltodextrin/gelatin, maltodextrin/chitosan and maltodextrin/β-cyclodextrin/arabic gum) on microencapsulation of plum phenolic compounds by spray drying. Encapsulation efficiency and storage stability of the microparticles were assessed. According to the authors, the stabilization of plum phenolic compounds was successfully achieved for all combinations of wall materials used. However, microcapsules produced with maltodextrin/chitosan showed higher retention of phenolics, above 94%, during storage for 60 days at 25 °C, compared to other microcapsules (between 80% and 90%) and to non-encapsulated phenolics (31%). Additionally, it was observed that the microcapsules with a smooth external surface demonstrated better protection of the phenolic compounds. Overall, the authors concluded that microcapsules produced with different wall materials improved the stability of phenolic compounds, presenting a potential for the commercial application of plum phenolics as nutraceutical products.

Natural and synthetic extracts of vanilla rich in polyphenols were microencapsulated by spray-drying using whey protein concentrate as a wall material [42]. The microcapsules produced from each core material were compared and analyzed for their physicochemical properties, antioxidant activity, and vanillin retention during storage. No significant differences were found in the physicochemical properties studied (moisture, water activity, color, hue angle, solubility, microencapsulation yield, bulk density) for either type of microcapsule produced. The microcapsules showed higher preservation of vanillin when compared to the non-encapsulated extracts during storage for 30 days at 25 °C, showing to be a good strategy for its stabilization. The authors concluded that with the results obtained it is possible to guarantee the maintenance of the antioxidant activity of the vanilla extract after microencapsulation, enabling their incorporation into functional foods.

Different living cells have been microencapsulated by spray-drying and successfully stabilized, such as *Saccharomyces cerevisiae* [43], *Bifidobacterium* spp. [45], *Lactobacillus casei* [46], and *Lactobacillus acidophilus* [44]. The feasibility of probiotics as they pass through the gastrointestinal system is an important step in the food field. Except for the last study, the main objective of probiotic microencapsulation was to evaluate the effect of different wall materials on the survival of probiotics during the in vitro simulation of gastrointestinal conditions. It was observed that the microencapsulation of probiotics improves the efficiency of bacterial survival, with the choice of the type of wall material being a decisive factor. Maltodextrin, arabic gum, whey protein concentrate, and gelatin were used as wall materials for *S. cerevisiae* by Arslan et al. [43]. Inulin and oligofructose were used to encapsulate goat's milk *Bifidobacterium*. Trehalose and maltodextrin were used by

Liao et al. [46] for skim milk *L. casei*. The authors of each study recommended, respectively, the use of gelatin or arabic gum, goat's milk itself or goat's milk/inulin, and skim milk itself as carriers of probiotic bacteria, providing a survival rate above 85% in the gastrointestinal tract.

Moreover, Reyes et al. [44] investigated the viability of microencapsulated *L. acidophilus* after microencapsulation process and during storage at 4 and 23 °C, under 10% and 97% vacuum. The wall materials used were maltodextrin, arabic gum, and maize starch. The viability of probiotic bacteria after microencapsulation reduced by on average 15% for all wall materials containing a count of approximately 8 log CFU/g. During the 60 days of storage, the combination of the temperature of 4 °C and 97% vacuum was shown to be more suitable to guarantee the minimum of 6 log CFU/g within the microcapsules, which meets the recommended levels to have therapeutic effects in the human host.

6. Incorporation of Microcapsules in Food Matrices

Quite a lot of studies have already been carried out on the microencapsulation of bioactive compounds and probiotics. However, there are few studies reported in the literature regarding how loaded microcapsules behave in terms of stability when incorporated in real food matrices [93]. An important factor regarding this incorporation is that food products should not be adversely affected, namely their sensory properties altered by the addition of microencapsulated bioactive ingredients [101]. Therefore, the incorporation of bioactive compounds into a food matrix presents a challenge for the food industry. Nevertheless, some good examples may be highlighted (Table 1).

Ursache et al. [21] microencapsulated carotenoids from sea buckthorn extract by coacervation using whey protein isolate and arabic gum as wall materials. The encapsulated powders obtained were incorporated into muffins in order to evaluate the influence of the food matrix on the stability of the bioactive compounds. The amount of total carotenoids, antioxidant activity, physicochemical characteristics, sensory analysis, and stability of the bioactive materials during storage of the food matrix were tested. The stability analysis of the encapsulated carotenoids incorporated into the muffins during storage at 25 °C for 21 days showed a loss of carotenoid content of approximately 55%. However, no significant changes in the color of the muffins were observed. The sensory analysis showed that the muffins containing microcapsules were preferred by the panellists, and neither aroma nor flavor of the sea buckthorn berries was detected.

Carotenoid-rich gac oil was microencapsulated in a mixture of whey protein concentrate and arabic gum (7:3) by spray drying [34]. The microencapsulated gac oil obtained was incorporated into a viscous food (yogurt), a low viscosity liquid food (pasteurized milk), and a dry food (cake mix) with the purpose of fortifying the food matrix. The authors analyzed these fortified products for color, peroxide value, and the amount of carotenoids present during four weeks of storage for yogurt and pasteurized milk, and during four months for cake mixing. Overall, they concluded that encapsulated gac oil can be successfully incorporated into these food products, exhibiting a slight color change and high levels of carotenoids during storage. However, further research is needed to study the release and bioavailability of carotenoids reached in the bloodstream after ingestion.

Green tea extract rich in polyphenols was microencapsulated in maltodextrin by spray-drying and freeze-drying and then added to bread [48]. This study evaluated the impact of microcapsules on bread quality and polyphenol content after baking. The amount of polyphenols present in the baked bread was 33% lower than in the dough before baking. In terms of bread quality, no significant differences were observed in the color or flavor of the breads or in the volume and crumb firmness between the samples incorporated with particles produced by spray-drying and freeze-drying. According to the authors, the fortification of bread with polyphenols of green tea maintains bread quality and most of the functionality of the bioactive compounds, suggesting that such fortification could be efficient to increase polyphenol intake.

Oancea et al. [15] studied the effect of microencapsulated anthocyanins incorporated into fermented milk to act as a prebiotic for *L. casei*. Microencapsulation of anthocyanins from an extract of sour cherry skin was performed by coacervation followed by freeze-drying using whey protein isolate

and arabic gum as wall materials. The microencapsulated bioactive compounds exerted the prebiotic function and favoured the growth of the *L. casei* probiotic during storage for 21 days. The number of viable cells in fortified samples remained in the 10^{10} CFU/g range.

7. Release of Encapsulated Bioactives from Microcapsules

One of the major challenges for the food industry is the development of effective systems for the controlled release of encapsulated bioactive compounds, which is dependent on the wall and core material types, microencapsulation method, particle morphology and size, and release conditions (e.g., solvent, pH, temperature) [23].

The phenolic compounds from grape marc extract were microencapsulated by spray-drying technology using maltodextrin, whey protein isolate, and pea protein isolate as wall materials [41]. The influence of the type of wall material on the release of total phenolic compounds and anthocyanins from the microparticles was evaluated under simulated gastrointestinal conditions over a period of 3 h. The release profile was similar for both simulated digestive fluids (gastric and intestinal). Protein-containing particles, especially with whey protein isolate, showed a slower release rate than other studied wall materials (maltodextrin and pea protein isolate). The authors report that the slower release of the phenolic compounds may be due to the smoother surface of the microparticles imparting a smaller contact area with the medium.

Medina-Torres et al. [37] evaluated the release of bioactive compounds from microcapsules of maltodextrin loaded with laurel infusions produced by spray-drying. Three different drying inlet temperatures (140, 160, and 180 °C) and two feed rates to the dryer (8 and 10 mL·min^{-1}) were used to produce the microcapsules. Both the feed rate and the drying temperature influenced the release of the core material in water (T = 37 °C, pH around 6). Microcapsules dried at 160 °C with a feed rate of 8 mL·min^{-1} had greater and more prolonged release of the core material. Approximately 70% of the bioactive compounds were released within 48 hours, which according to the authors suggests a good absorption of the polyphenols in the small intestine.

The release rate of microencapsulated lime essential oil by spray-drying using mixtures of whey protein concentrate with different maltodextrin equivalent dextrose (5, 10, and 20 DE) was evaluated by Campelo et al. [22]. The release of the essential oil was studied in mineral oil at 25 and 45 °C. The percentage of the encapsulated core material that was released was less than 75% in both conditions. However, a greater essential oil release was observed with the increase of the degree of dextrose equivalent

Dima et al. [23] studied the release mechanism of microencapsulated coriander essential oil by spray-drying. Alginate, chitosan, chitosan/alginate, and chitosan/inulin were used as wall materials. In this study, in vitro release of essential-oil-loaded microparticles simulating pH and temperature (37 °C) conditions of the gastrointestinal tract and food product processing (65 °C) was evaluated. The results showed that the release of the essential oil from the particles was influenced by pH and temperature. For example, in an acidic fluid (pH 2.5) at 37 °C, there was a higher release rate from the chitosan particles. They showed a rapid release rate in the first 60 min, releasing almost 60% of the essential oil that was encapsulated. Thereafter, the release slowed to a peak of 74.5% after 120 min. In contrast, the alginate particles presented a lower release under the same conditions, almost 29% after 120 min. This difference in the release of the essential oil from the microparticles was justified by the low degree of swelling of the alginate microparticles and their rigid polymeric chains, which hindered the release by diffusion. In terms of temperature, as the temperature increased to 65 °C, the rate of release of the essential oil also increased for both pH values.

The release profile of microparticles loaded with carotenoid-rich palm oil incorporated into food matrices was evaluated by Rutz et al. [53]. The microparticles were produced by a complex coacervation method using chitosan and xanthan gum as wall materials, and were then incorporated into yogurt and bread. Food samples with microencapsulated bioactive compounds were placed under conditions simulating the gastrointestinal tract. For the bread, 38.9% of the carotenoids were released,

while for the yogurt 50.1% were released. Additionally, according to the results obtained in the study, before incorporation into food, the bioactive materials from the microparticles had greater release during the simulation of the gastrointestinal tract, with greater degradation. The authors inferred that the interaction of food matrices with microcapsules may protect the encapsulated bioactives, suggesting that the loaded microcapsules produced have great potential to be incorporated into foods, mainly yogurt.

8. Conclusions and Future Perspectives

An overview of the latest studies regarding the application of microcapsules in the stabilization of functional compounds for food applications is presented. There has been growing interest in the development of microcapsules loaded with probiotic bacteria, phenolic compounds, and carotenoids from natural sources. The main objectives of this strategy are to improve the stability of bioactive compounds against adverse environmental conditions, their incorporation into food matrices conferring functional properties to food products, and to enable their controlled release at a specific target of the gastrointestinal tract after food ingestion. The most widely used encapsulating processes are spray-drying followed by complex coacervation. A fine-tuned optimization of process conditions is required, including the selection of the wall material for a specific core material, to produce loaded microcapsules with suitable characteristics for a specific end application.

Although several functional compounds have already been successfully stabilized by microencapsulation, there is still a lack of knowledge regarding how loaded microcapsules behave in terms of release rate when added to complex food matrices. Additionally, further studies are needed to access the effective bioavailability of released functional compounds after ingestion, which is correlated to the health benefits claimed for most of those compounds.

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